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PRODUCT DUST AND AEROSOL MONITORING PROGRESS REPORT.

Alfred Pfanstiehl.

July 18, 1944.

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ABSTRACT.

Dry filter paper has been shown to serve as an adequate collector of particulate matter from the air as the basis of a monitoring device. An ion chamber is arranged to receive alpha particles from the collected active material as air is pulled through the paper. This, together with the associated electronic circuit with a meter to read the accumulated activity and an alarm buzzer to warn when a certain level is reached, is known as the "Sneezy" product air monitor. The collection of short-lived emitters along with product particles has been shown in preliminary tests to produce less ionization than that due to a dangerous level of product concentration.

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I. OVERVIEW.

The development of "Sneezy" to monitor for particulate alpha active dusts and aerosols involves a study of filter paper efficiency and short-lived activity which is collected along with the dangerous long-lived product. This work, although not completed, will be summarized as well as the associated electronics development.

"Sneezy" is an adaptation of the same ion chamber arrangement which has proved successful in the "Pluto" surface alpha contamination indicator. It combines both high and low level continuous monitoring. First, it collects and integrates the particulate matter from the same air breathed by the protected personnel, and in the event of an accident sounds an alarm when it has "inhaled" a certain small fraction of what is considered a lifetime dose of product; and second, it provides papers which can be counted in a special alpha chamber as a daily check on the general level of air contamination for long-term safe working conditions.

This report will review the original problem put to the Instrument Section; give the reasoning and mathematics behind the development, with some special notes on filter paper action and data on observed short-lived alpha activity; and describe the instrument and the present knowledge of its sensitivity and operational factors.

II. REQUIREMENTS.

In the original request from Site "Y", two types of instruments were thought necessary: one to warn definitely that an accident had occurred whereby the level of product contamination in the air had reached 0.05 alphas/sec/cc. and the other to indicate when the general level exceeds  $5 \times 10^{-6}$  alphas/sec/cc. (See appendix for calculations by which these figures were obtained.)

Since the low level figure is in the same ionization range as that of cosmic rays and the natural short life alpha emitters (radon), some means of collection and concentration of the product is obviously called for. The time factor is relatively unimportant, as the check need be only on a daily basis.

The high level, however, requires continuous monitoring. The original idea was to have an instrument sound an alarm if some accident caused the concentration to rise to 0.05 alphas/sec/cc., at which a man could get a "lifetime dose" in one-half hour. Calculations showed this might possibly be done with a 10 liter chamber. That this would be inadequate was readily apparent; what if the level was only one-half of this? A man might get over his "lifetime dose" in several hours and not know it until the results of the periodic (perhaps daily) low level check were in. Continuous integration and indication of the total accumulated activity is clearly necessary, with an alarm set to go off at a certain level. Then, by noting the rising speed of the activity, the worker can gauge the actual danger present and act accordingly. If he sees the needle rising noticeably fast, his immediate exit, while holding his breath, would be required.

The "lifetime dose" and "tolerance" calculations, given in the appendix, are quite hypothetical but represent an agreement at least in the general order of magnitude, of the workers in this field. The sensitivity of "Sneezy" was designed to exceed these tentative levels by a clear margin. A thorough testing program is in progress, and from the preliminary results it appears that to date this is the best product air monitoring instrument in evidence.

### III. COLLECTION METHODS CONSIDERED.

Before deciding on the use of filter paper as a collector, these methods were investigated and rejected for the reasons given:

1. Electrostatic precipitator. There is little real evidence of the efficiency of this method, particularly for the smaller sized particles. The powerful electric field certainly sets up forces, "electrical wind", tending to drive out the dust, but tests show that at practical rates of air flow much material can be missed. The complications of high voltage supply, careful design and operation, and the infeasibility of combining the continuous reading, integrating warning instrument with the low level indicator ruled out this method. It is the opinion of Dr. Rodebush, of the NDRC, Project #10, and of Dr. Hodge, of the Rochester Project, both of whom have been working with dusts and filtration problems for some time, that the electrostatic precipitator is generally not as efficient as filter paper.

2. Thermal precipitator. This is apparently still a laboratory device, useful only in taking out particles for size and distribution determinations. The rates of flow used are only several cubic centimeters per minute, which immediately rules it out, unless a faster collector could be devised.

3. Impinger. Long used in industrial dust measurements, the impinger has definite limits on the particle size collected, and does

Not concentrate the matter from large volumes of air. However, recent developments have greatly improved this principle by using rubber diaphragms, etc., and the suggestion has been made to combine an impinger and a fluorescent screen upon which actual scintillations might be counted. This has not been tried as yet.

4. Liquid bubblers. Respiratory project work has shown that efficient dust removal can be achieved if fine sintered glass discs and specially developed bubblers are used with known solvents for the particles in question. But a chemical method, requiring repeated tracer quantity analyses, does not sound encouraging, nor does it seem possible to have continuous monitoring.

5. Activated absorbers. If fumes are involved in the monitoring problem, we can benefit by the comprehensive work already performed on activated charcoal and other absorbers in the war gas protection projects.

#### IV. FILTER PAPER

Most of the hesitation to accept dry filter paper as an efficient dust collector is based on a false picture of its action. The mechanism of filter action has been thoroughly investigated by scientists who have developed the current combat gas masks in government projects. Dr. Rodebush, at the University of Illinois, informs us that generally the collection efficiency increases as the particle size becomes smaller, even down the order of 0.01 microns and less. With the new fine fiber papers, in the range of 0.05 to 1 micron the stopping power goes through a minimum, and for larger sizes, practically complete filtration is effected. Total efficiencies are often over 99%. This rather surprising function is explained as follows:

1. Only the largest particles (50 microns and more) are stopped by sieve action, which is of little importance.
2. Smooth, streamlined air flow around each fiber of the paper is shown to be the case at normal rates of flow.
3. The momentum of particles down to about 1 micron is such that they tend not to follow the curved path of air around each fiber, but to continue in their straighter paths to strike a fiber. It appears that surface forces exceed gravitational forces, so all particles that actually strike a fiber, adhere to it.
4. In the critical range, the particles do tend to follow the streamlines around the fibers, and, therefore, are less effectively stopped. This is in the range of about 0.5 to 1 micron diameter.
5. The smaller particles are subject to Brownian movement which drives them out of the air stream and on to the nearest fiber.

- 4 -

Small fiber size is important, so the air stream are as narrow as possible. This diffusion effect increases as the particles become smaller and more mobile, until other molecular forces become involved.

Special papers, made of newly developed extremely fine rayon, asbestos, and other fibers, are being manufactured for gas mask use. Well over 99% of all particulate material can be removed with several layers of these papers, which are rather lightly compressed for low air resistance.

Our problem does not concern air resistance, but rather with the location of active particles in the paper structure and the degree of absorption of the alpha particles. Special hard finished papers made of the new fine fibers (1 micron and less diameter) have been ordered. These will undoubtedly increase the efficiency, for the paper in use now is the standard Whatman 50, a hard finished cellulose paper with fibers of about 20 or 30 microns diameter. This paper has been tested (on Dr. Rodebush's standardized equipment) for particles of about 0.05 microns, where the retention was roughly 50%.

To learn how well the alpha particles from the active material embedded in the paper may be measured, and to see what effects the natural radioactivity might have, a series of 80 samplings were made. For each test, the place, time, flow rate, and total volume of air pulled through the paper were recorded. Usually about 1000 liters were filtered at 3 liters per minute through a paper 4.25 cm. diameter. This corresponds to a linear flow rate of about 4 cm/sec. which is within the range of linear rates used in other studies of filter paper and in the "Sneezy" chamber. Figures 1 and 2 show one of the simple filter paper holders and flow meters used. The paper diameter was chosen as a standard available size that would fit in the regular alpha chambers used in the analytical work. Whatman 50 paper was found to have the lowest background count and was the hardest finish available.

Dust samples were taken in various laboratories of the New Chemistry building and also in other buildings where no product or other radioactive chemistry was performed. Often two papers were run in parallel from the same pump. The papers were put in an alpha chamber and counted immediately at the end of the collection period. Ten minute counting periods were generally used because of the low counts. The decay curve was followed in many of the tests. A number of days after the collection (when the sustained activity had been established) each paper was chemically digested in nitric acid and a lanthanum fluoride coprecipitation performed to separate out the product, if present. This chemical analysis was carefully checked with known tracer quantities and parallel blanks with each set of papers.

The results of this work is summarized as follows:

1. Two clear decay periods are evident in nearly all of the tests.

Fast: About 30 to 60 minute half-life which usually carries activity down to about 10% of the initial activity in 2½ hours, leveling off at about 5% in 5 hours.

Slow: 1 to 3 days half-life, carrying activity on down to permanent level in 4 to 6 days. (Further decrease in activity may be due to the handling of the papers.)

See Figure 3, and its key, for examples of these curves.

2. The initial activity (at 0 time, when collection is stopped and counting begun) seems to depend somewhat on the location of the test and perhaps the weather. Parallel tests on the same days show excellent duplication, but tests of equal quantities of air in the same rooms on different days are widely divergent. This initial, fast-decaying alpha activity could be due principally to Ra A (3 min.) and Ra C (19.5 min.), with Ra B (26.8 min. beta) determining the main decay rate.

In the non-air-conditioned rooms when these tests were made, initial counts in the neighborhood of 300 c/min. for 1000 liters were occasionally found. This is equivalent to  $5 \times 10^{-6}$  alpha/sec/cc, which is  $10^{-4}$  of the calculated high level danger point of product contamination. Most of the tests showed much lower initial activities--usually about  $10^{-6}$  alpha/sec/cc.

3. Permanent activity appeared in a few of the tests, and was attributed to the product collected. There was no correlation between the permanent and the initial activities and its sporadic occurrence indicates that the product dusts are quite localized in space and time, and do not diffuse to a general concentration in the air.

4. The chemical digestion and separation of the product indicates that a direct count from the paper is well within the correct order of magnitude. Corrections for loss in the chemical work were made with known tracer quantities, yet in the 8 papers found with sustained activity, the digestion process yielded up to 40% less count. Perhaps some of the apparently "permanent" activity was due to other relatively long-lived emitters, since the product coprecipitation is quite selective. All this work was necessarily on a tracer scale, where quantitative results are difficult.

5. The data on 60 tests breaks down as follows:

Initial Activity (extrapolated to 0 time--the end of the collection period, long life activity subtracted, and count normalized to that for 1000 liters air.)

In Product Labs. (Total Tests = 45)		In Hallway and Shop Previously Used as Metal Storeroom. (Total Tests = 5)		In Other Places- No Contamination. (Total Tests = 10)	
Number of Tests	$\alpha$ /Min.	Number of Tests	$\alpha$ /Min.	Number of Tests	$\alpha$ /Min.
10	0-20	1	0-20	6	0-20
8	21-40	0	21-40	3	21-40
6	41-60	0	41-60	1	41-60
5	61-80	0	61-80	0	61-80
6	81-100	0	81-100	0	81-100
10	100-600	4	100-600	0	100-600

Sustained Activity (after 5 days) and results of digestion and co-precipitation.

Note: Long life activity occurred only among those tests made in labs doing product chemistry. All others decayed to 3 c/min or less, with counts after digestion also 3 or less. Each of the 8 tests with long-life activity is here reviewed. A "geometry" of 25% has been assumed for the  $\alpha$ /sec/cc and the curies/cc calculations.

Air Flow Rate Liters/ Min.	Total Liters of Air Filtered.	$\alpha$ /Min. Direct From Paper After Five Days.	$\alpha$ /Min. After Di- gestion.	$\alpha$ /sec/cc at 25% Geometry.	Equivalent curies/cc.	Remarks.
3	1350	38	77	$3.9 \times 10^{-6}$	$1.1 \times 10^{-16}$	Parallel Test
1	450	21	20	$3.1 \times 10^{-6}$	$8.4 \times 10^{-17}$	
3	990	450	-	$3.0 \times 10^{-5}$	$8.4 \times 10^{-16}$	Highest Found.
2.5	775	13	7	$1.1 \times 10^{-6}$	$3.0 \times 10^{-17}$	Parallel Test
2.5	775	28	20	$2.4 \times 10^{-6}$	$6.4 \times 10^{-17}$	
3	1305	8	7	$4 \times 10^{-7}$	$1.1 \times 10^{-17}$	
2	860	8	5	$6 \times 10^{-7}$	$1.6 \times 10^{-17}$	
3	990	17	11	$1.1 \times 10^{-6}$	$3.0 \times 10^{-17}$	

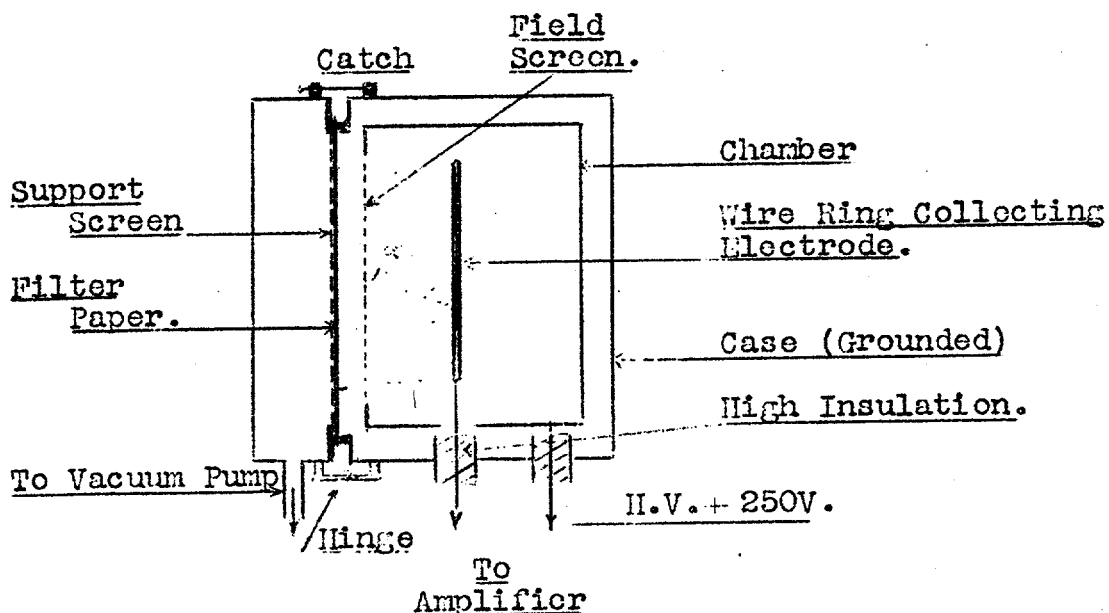


Further work is planned along these lines with the new papers and with a variety of alpha active dusts under more controlled conditions. The presence of a fair amount of city dust, for example, which definitely collected on the top surface of the paper, may have affected the results; the building was not air-cleaned when the tests were run.

The indications at present are excellent that a sensitivity well within the probable degree of question of the tolerance calculations has been achieved, even with the definitely inefficient ordinary papers now used. The new papers will have much faster air flow and greater retentivity. (For more details on the work by Dr. Rodebush, see the report WPJ-24, May 27, 1944, which has the Chicago Laboratory Library Number N-1172.)

#### V. "SHIELDY" CHAMBER.

The chamber is a simple open screen ion chamber mounted directly in front of a 12.5 cm paper through which the air is pulled. It is connected by a hose to the pump and by cables to the chassis containing the power supply and amplifier, and can be placed in any position where it can "inhale" the same air as the personnel, or in the ventilation exhaust ducts. In the figure is shown a cross-section sketch of its design:



Air inlet slots around the wall of the external cylinder are located at the screen's edge (See Figure 4). Thus, active dust particles are collected on the paper, where a portion of their alphas can travel into the ionization chamber. The re-

latively negative collecting ring attracts the positive ions formed, and this current is measured as the voltage drop across a high resistor to ground.

#### VI. "SNEEZY" CIRCUIT.

Simplicity and dependability, together with the maximum practical sensitivity were the considerations in developing the circuit used. With the use of careful drying and high insulation varnish on the input switch, resistor, tube, and socket, a 1011 ohm resistor can be used on the input. The voltage developed by the ion current through this resistance is on the #3 grid (lowest grid current) of a 959 tube, whose plate is directly coupled to the #1 grid of a 1A5. A 0-100 microampere meter is used as a voltmeter to read the change in plate voltage of this tube, which is then a function of the chamber ion current. A 6SJ7 tube operates the alarm relay at any desired level. The rest of the circuit consists of a regulated power supply and the necessary adjusting potentiometers. A sensitivity check input is provided by which a voltage between 0 and 1 volt may be used to replace the chamber and resistor. The only external controls are a "SET" and "READ" switch, a "ZERO SET" and a "RELAY SET". The voltage sensitivity of this amplifier is in the neighborhood of 0.6 volts full scale.

See the appendix for a calculation of the required ion current and resultant sensitivity to alpha activity. Development details are contained in the Report #CP-1002 by W. Hinch. Manufacturing details on this circuit may be obtained from Report #AD-50234, Engineering Group, May 29, 1944. Several small changes have been made since this date, however. This circuit has not proved as stable as one might wish, and a new circuit, using more recent tubes, is in the development stage.

#### VII. OPERATIONAL NOTES.

The placement of the collecting head and chamber is a matter requiring more experience. Indications so far have been that product contamination is a highly localized condition rather than a general concentration at a definite level. It appears that a centrifuge, a fuming beaker, or a dry transfer, for instance, may release a considerable amount of product dust in the air which, if in particles upward of 1 micron in size, would rather quickly settle out and adhere firmly to the bench tops or floors. Smaller particles would tend to remain air-borne and therefore constitute the major problem. This is in accord with Stokes law of fall.

It is obviously important that the "Sneezy" chamber be placed in the places where the highest possible contamination is encountered in the air breathed by the personnel. It will probably always be possible for a man to get a concentrated whiff of dust that "Sneezy" misses, or vice-versa, but this chance must

be minimized. Ventilation exit ducts would appear to be good places, also.

Open flames, steam, and common dust will occasionally cause ionization in the chamber. Weather conditions also seem to effect the readings somewhat. These effects have not been noted to approach the alarm level, however, and are always temporary. Some natural activity is also accumulated in long runs, but this also has not risen high enough to operate the alarm.

A daily routine should be established of changing the paper, checking the zero set, performing a quick check on amplifier sensitivity with a battery volt-box, and checking the chamber insulators with a standard uranium source placed in lieu of the paper. (Dust or moisture on this insulator could reduce the sensitivity of the instrument.) The papers can be allowed to decay for about 24 hours or more, if necessary, and then counted in a special alpha chamber, as being developed at Chicago. Permanent alpha activity can be attributed to product, and thus the long-term contamination level ascertained.

#### VIII. CONCLUSIONS

Work is in progress to obtain more data on the special efficiency of filter papers for this instrument, and the new, fine-fiber paper is ordered. Meanwhile, the conclusions reached by other projects making comprehensive study of filter action have been accepted in the development of "Sneezy".

As with any new instrument, "Sneezy" must be put into widespread service with a careful record kept of all operating conditions so that corrections and improvements may be made, before forming a final estimate of its usefulness.

All credit for the electronics and original integrating chamber design goes to Mr. E. Holloy, Mr. W. Hinch and their associates, while the filter paper efficiency and natural activity study is being done by Mr. A. Pfanstiehl, all of the Instrument Section at Chicago. Dr. J. J. Nickson of the Health Group has been a close consultant, and credit is due Mr. R. Patton of the Chemistry Section for his early work and help on the filter paper work. The chemistry was done by Mr. Beard and Mr. Britton.

APPENDIX.

For uniformity, the calculations for concentrations have been reduced to alphas/sec/cc. Conversions to curies and to mass of product may be made by multiplying by those factors or their reciprocal, which are equal to 1, according to the proper cancellation of units:

$$\frac{1 \text{ curie}}{3.7 \times 10^{10} \text{ alphas/sec}} = 1$$

$$\frac{1 \text{ gm product}}{2.3 \times 10^9 \text{ alphas/sec product}} = 1$$

(For 100% geometry,  $10^{-6}$  gm  $\Lambda$  = 140,000 alphas/min = 2,300 alphas/sec.) 5 micrograms  $\Lambda$  is considered a tolerance "lifetime dose".

Breathing Rate.

Approximate volume of air in alveoli per breath = 500 cc/breath.  
At 18 breaths/minute = 9 liters/minute = 540 liters/hour = 150 cc/second. On a 48 hour per week basis, the approximate total volume breathed over a two year period

$$V = 2.6 \times 10^9 \text{ cc/2 yrs.}$$

Long Term Level

So, to get 5 micrograms in 2 years, the concentration would be

$$\frac{2 \text{ yrs.}}{2.6 \times 10^9 \text{ cc}} \times \frac{5 \times 10^{-6} \text{ gm}}{2 \text{ Yrs.}} = 2 \times 10^{-15} \frac{\text{gm}}{\text{cc}}$$

Assuming all dust reaching alveoli is absorbed. This is a very generous assumption.

This reduces to

$$2 \times 10^{-15} \frac{\text{gm product}}{\text{cc}} \times \frac{2.3 \times 10^9 \text{ alphas/sec}}{1 \text{ gm product}} = 5 \times 10^{-6} \frac{\text{alphas/sec}}{\text{cc}}$$

This is about the same activity as that due to the natural radon concentration. A questionable amount of radon is apparently concentrated on the paper fibers or dust particles, but it is short-lived. Other short-lived activity also makes it necessary to wait before counting the paper to determine this low level.

### High Level

If the concentration gets so high one could get 5 micrograms in one-half hour, this calculation is made:

$$\text{Total volume of air} = \frac{150 \text{ cc}}{\text{sec}} \times 1800 \text{ sec} = 2.7 \times 10^5 \text{ cc.}$$

$$\frac{5 \times 10^{-6} \text{ gm}}{2.7 \times 10^5 \text{ cc}} \times \frac{2.3 \times 10^9 \text{ alphas/sec}}{1 \text{ gm product}} = 0.05 \text{ alphas/sec/cc.}$$

### "Sneezy" Theory

The high level figure appeared rather too high for comfort, so "Sneezy" was calculated to be at least several times as sensitive.

With a voltage sensitivity of 0.5 volts and a  $10^{11}$  ohm resistor, the necessary ion current is

$$I = \frac{0.5 \text{ volts}}{10^{11} \text{ ohms}} = 5 \times 10^{-12} \text{ amperes}$$

The approximate number of ion pairs formed in the chamber per alpha particle =  $10^5$  i.p.

Then,

$$5 \times 10^{-12} \frac{\text{coulombs}}{\text{sec}} \times \frac{1 \text{ i.p.}}{1.6 \times 10^{-19} \text{ coulomb}} \times \frac{1 \text{ alpha}}{10^5 \text{ i.p.}} = 300 \frac{\text{alphas}}{\text{sec}}$$

Assuming about 50% loss and absorption in the filter paper and a 25% geometry of the chamber, the required activity becomes

$$\frac{300}{0.125} = 2400 \frac{\text{alphas}}{\text{sec.}} \quad \text{for a full scale reading. The alarm can}$$

be set to go off at  $\frac{1}{4}$  scale, so about 600  $\frac{\text{alphas}}{\text{sec}}$  accumulated activity is required.

With the papers and pumps in use at present, about 60 liters/minute of air is filtered, which is approximately 6 times the normal respiration rate. This is  $10^3 \frac{\text{cc}}{\text{sec}}$ , so if the alarm goes off in 1 minute, when 600 alphas/sec have accumulated, the concentration was

$$\frac{600 \text{ alphas/sec}}{10^3 \text{ cc/sec} \times 60 \text{ sec}} = 0.01 \text{ alphas/sec/cc. in which one might}$$

get a "lifetime dose" by staying in the room for 2½ hours. If he left in one minute, he would have gotten about 0.013 of a "lifetime dose".

If it took an hour to accumulate enough to set off the alarm, the concentration was

$$\frac{600 \text{ alphas/sec}}{10^3 \text{ cc/sec} \times 3600 \text{ sec}} = 0.0002 \text{ alphas/sec/cc}$$

in which one might get a "lifetime dose" in 125 hours, or only 0.0031 of this if he left in one minute after hearing the alarm.

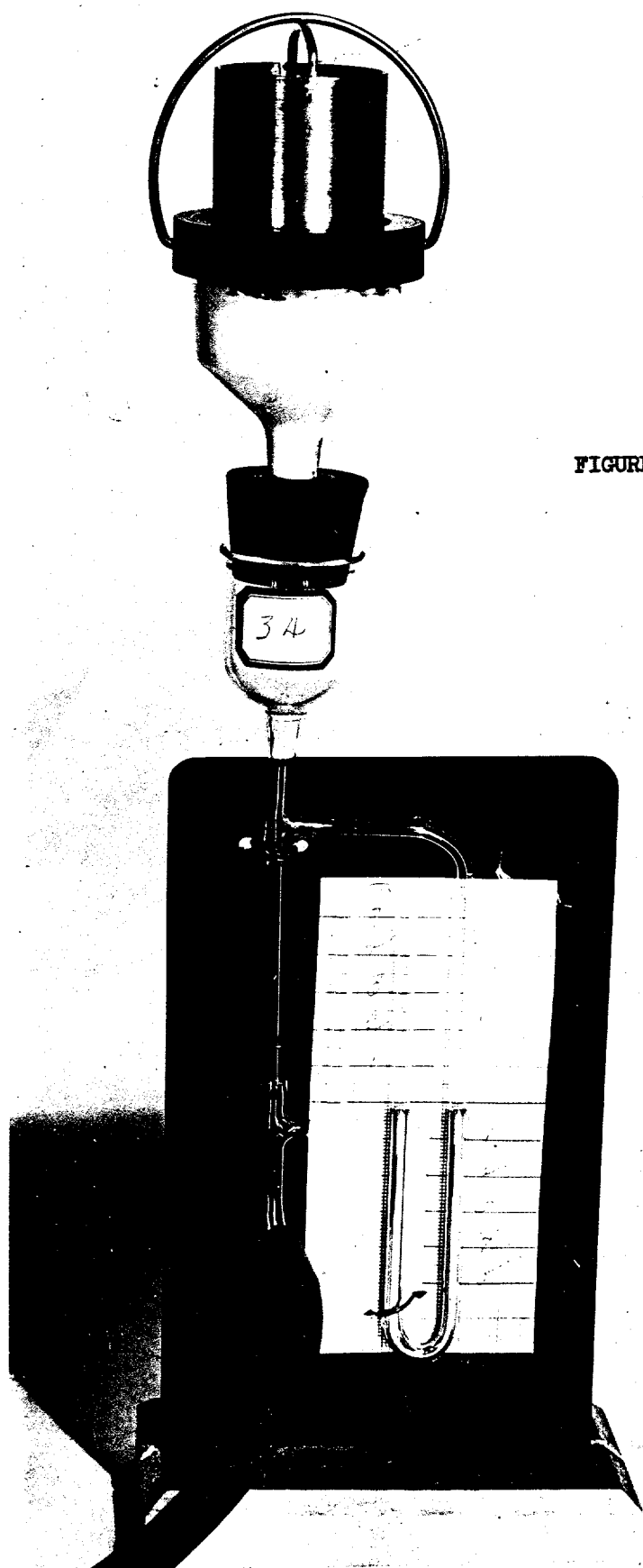
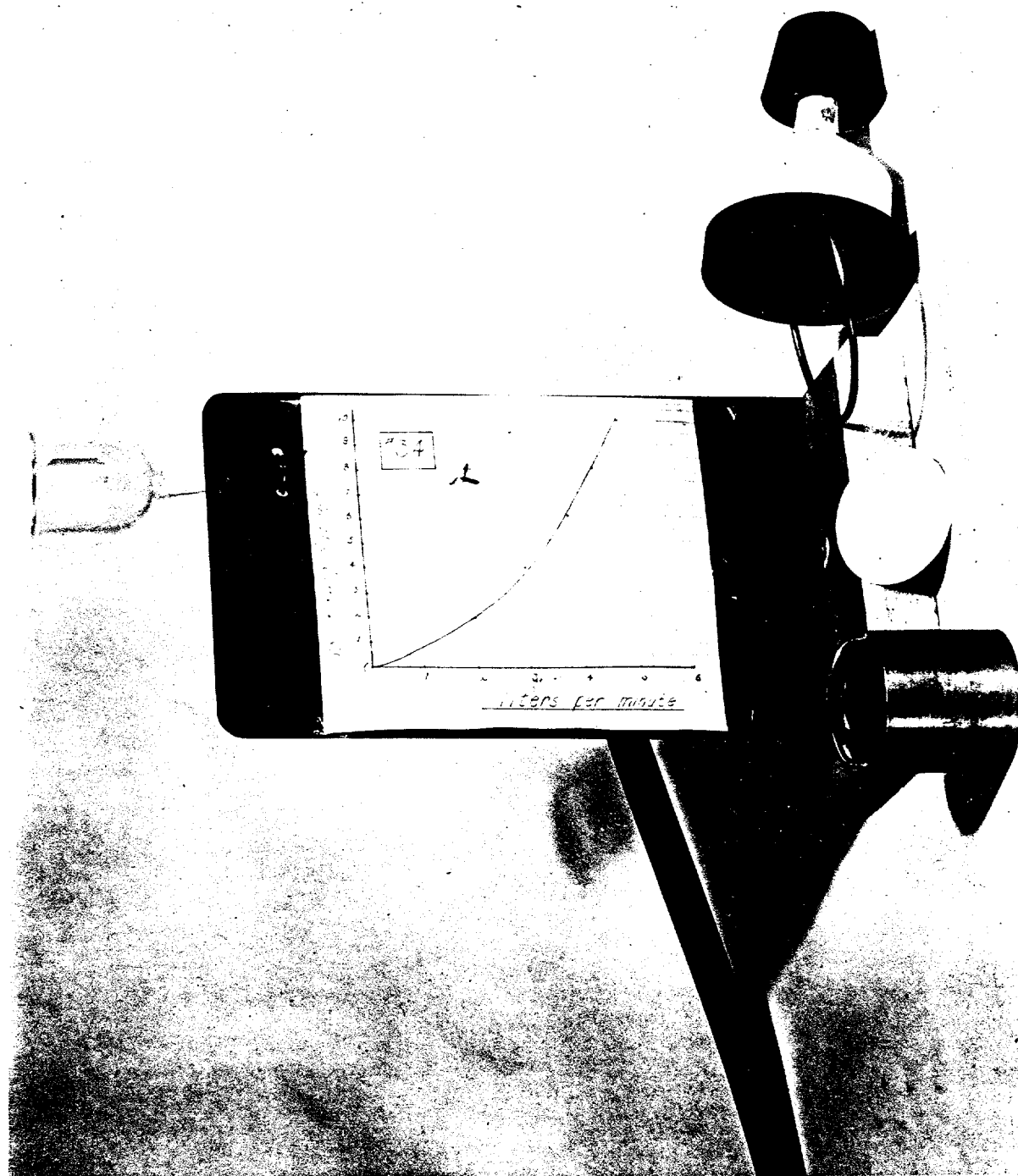


FIGURE 1.

FIGURE 2

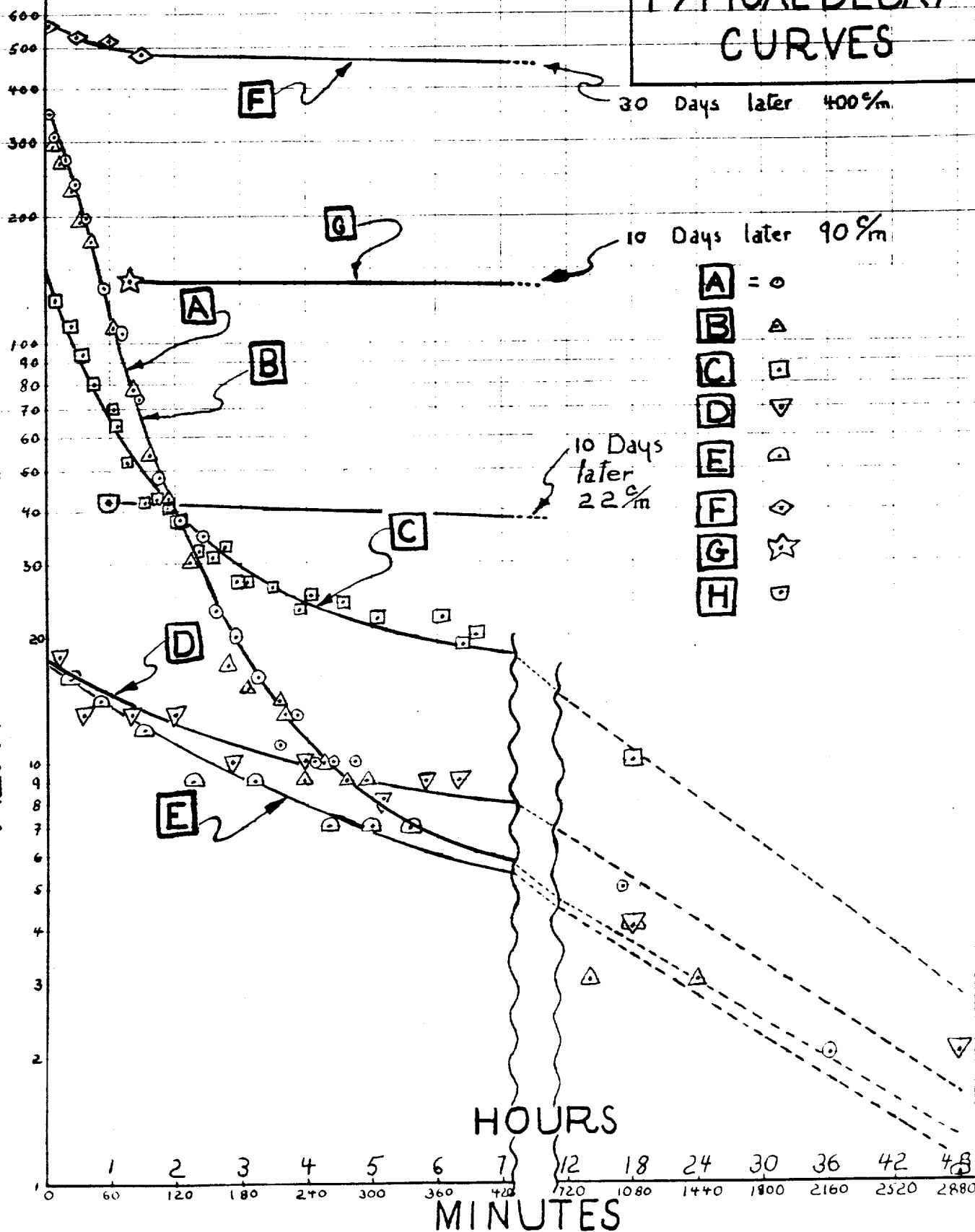




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FIGURE 3  
TYPICAL DECAY  
CURVES

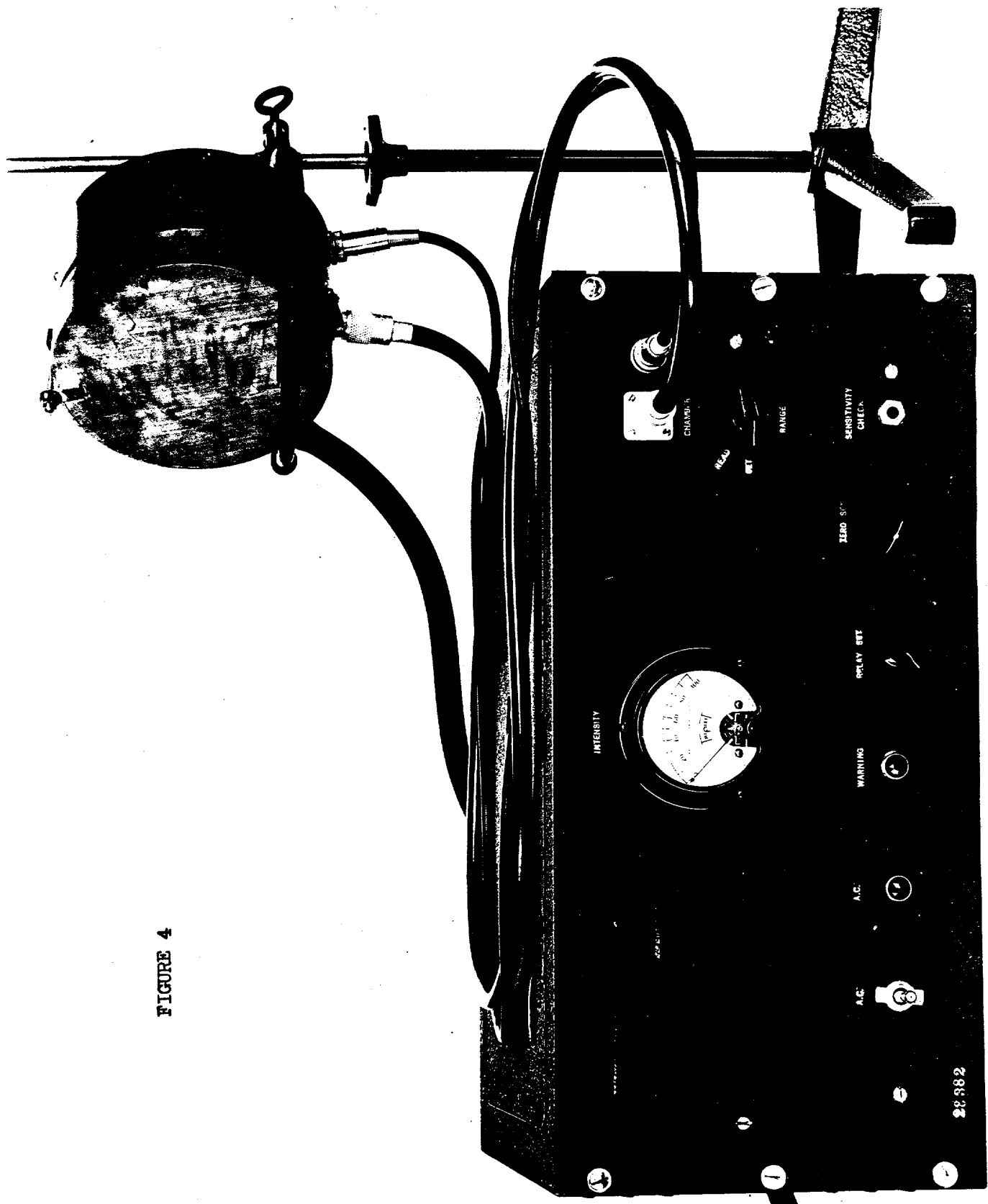
ALPHA COUNTS PER MINUTE



KEY - FIGURE 3

CURVE	A	B	C	D	E	F	G	H
ROOM	In Hallway near 19B, where previously much U metal was stored. Much traffic stirring up dust.		NC 10	Ryerson 358		NC 10	NC 9	
TYPE OF WORK			Product Chemistry	ELECTRONICS No $\propto$ Work.		Product Chemistry	PRODUCT CHEMISTRY	
Total Liters of Air.	1140	1140	1260	9400	9400	990	1350	450
Remarks	Parallel test - filters adjacent and at same flow rate. Note accurate duplication. No sustained activity.		Same position as for F. collected no sustained activity.	Equilibrium reached; 10 times as much air as in a previous test, but less than two times initial activity.		Highest sustained activity obtained.	Three times air volume yielded about three times activity. Tests were adjacent.	

FIGURE 4



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